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AUTHOR(S):

Chen, Jun; Itakura, Ryuji; Nakajima, Takashi

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Reconstruction of attosecond pulses using two-color pumping

Jun Chen,^{1,2} Ryuji Itakura,³ and Takashi Nakajima^{2,*}

¹College of Optical and Electronic Technology, China Jiliang University, Hangzhou 310018, China

²Institute of Advanced Energy, Kyoto University, Gokasho, Uji, Kyoto 611-0011, Japan

³Kansai Photon Science Institute, Japan Atomic Energy Agency, 8-1-7 Umemidai, Kizugawa, Kyoto 619-0215, Japan

*Corresponding author: t-nakajima@iae.kyoto-u.ac.jp

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We propose a two-color pumping scheme to characterize attosecond extreme ultraviolet (XUV) pulses. The fundamental and its second harmonic of a femtosecond Ti:sapphire laser create a coherent superposition of the $4p$ and $5p$ states of K, and we retrieve the spectral phase of the XUV pulse from the phase offset of the photoelectron signal as a function of time delay after the pump pulse. The scheme is technically simple and efficient to characterize ~ 100 as pulses. © 2011 Optical Society of America

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1. INTRODUCTION

The generation of attosecond pulses has attracted much attention as a novel light source [1–3]. Because the use of attosecond pulses enables us to reach the unprecedented temporal resolution as demonstrated in recent works [4,5], an accurate characterization of attosecond pulses is crucial for any application. Although there are a few known schemes, such as RABBIT, which requires the atomic phases in advance to determine the temporal structure of the individual attosecond burst [6,7], FROG-CRAB, which relies on strong field approximation (SFA) to retrieve the temporal profile of the attosecond pulse [8–11], and others [12,13], an alternative scheme is still highly desired to check the reliability and/or the accuracy of pulse reconstruction.

Along this line a photoelectron SPIDER using a fine-structure doublet of the Cs atom has been proposed (Cs SPIDER) [14]. It was demonstrated that the photoelectron signal induced by the time-delayed XUV pulse after the pump pulse provides sufficient information on the spectral phase by the standard SPIDER algorithm. Since it is purely a perturbative method and the spectral phase is extracted from the phase differences as a function of frequency, the absolute value of the atomic phase is not needed. This means that an accurate reconstruction of the attosecond pulse is possible without any approximations. The problem of the Cs SPIDER, however, is that it cannot be used for the reconstruction of the attosecond pulse since the energy shear is too small, 0.069 eV. Moreover the pump wavelength needed for the Cs SPIDER is 850–890 nm, which is not very convenient if one employs a Ti:sapphire laser tuned at ~ 800 nm. If, instead, two electronic states with a >1 eV energy separation are coherently excited by the vacuum ultraviolet (VUV) pump pulse with a few femtosecond duration (VUV SPIDER), an XUV pulse down to ~ 100 as can be characterized [15]. Still, the remaining difficulty is the need of an intense VUV pump pulse with a few femtosecond duration.

In this paper we propose a further improved version of photoelectron SPIDER using a two-color pumping (two-color

SPIDER), in which the fundamental (797 nm) and its second harmonic of a Ti:sapphire laser create a coherent superposition of $4p$ and $5p$ of the K atom. There are two main advantages for the two-color SPIDER scheme. First, we can easily obtain a large energy shear, 1.45 eV, which is necessary to characterize pulses with a sub-100 as duration. Second, we can easily pump the large portion of the atoms, say $\sim 50\%$, to the excited states, and, hence, the magnitude of the photoelectron signal is comparable with that of, say, FROG-CRAB where ionization is directly induced from the initial (ground) state. Since none of the photoelectron SPIDER schemes mentioned above utilizes any approximations, such as SFA, and in particular the two-color SPIDER is capable of characterizing XUV pulses down to sub-100 as with a sufficient signal intensity, it can be an alternative method to characterize attosecond XUV pulses.

2. DESCRIPTION

The scheme we propose is shown in Fig. 1. The level structure of the K atom best matches the use of the fundamental (797 nm) and its second harmonic of a Ti:sapphire laser for the two-color pumping to create a coherent superposition of $4p$ (labeled as $|1\rangle$) and $5p$ (labeled as $|2\rangle$) from the $4s$ (labeled as $|g\rangle$) ground state. Note that the purpose of these pump pulses is to prepare a coherent superposition of $4p$ and $5p$ states, and, hence, there can be a time lag between them as long as the time lag remains constant during the entire measurement. At a time delay τ after the two-color pump the attosecond XUV pulse is turned on to induce photoionization. The XUV pulse we would like to characterize is defined in the frequency domain as $E(\omega^{\text{XUV}}) = \sqrt{I(\omega^{\text{XUV}})} \exp[i\phi(\omega^{\text{XUV}})]$. Assuming that all three pulses are linearly polarized along the same direction, we can derive the expression for the photoelectron angular distribution at the photoelectron energy ε and time delay τ . If we define the photoelectron ejection angle, Θ , with respect to the polarization angle, it reads [15,16]

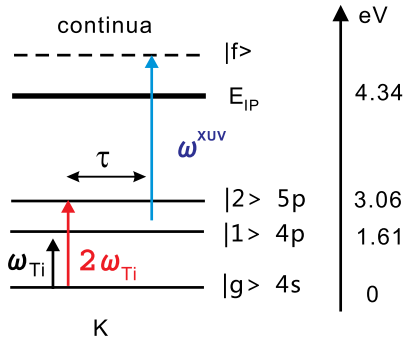


Fig. 1. (Color online) The two-color SPIDER scheme. The energy spacings of $4s-4p$ and $4s-5p$ fit the use of the fundamental (797 nm) and its second harmonic of a Ti:sapphire laser.

$$S(\epsilon, \Theta, \tau) = S_{gg}(\epsilon, \Theta) + S_{11}(\epsilon, \Theta) + S_{22}(\epsilon, \Theta) + S_{1g}(\epsilon, \Theta, \tau) + S_{2g}(\epsilon, \Theta, \tau) + S_{21}(\epsilon, \Theta, \tau), \quad (1)$$

where $S_{gg}(\epsilon, \Theta)$, $S_{11}(\epsilon, \Theta)$, and $S_{22}(\epsilon, \Theta)$ represent the photoelectron signals from states $|g\rangle$, $|1\rangle$, and $|2\rangle$, respectively. $S_{1g}(\epsilon, \Theta, \tau)$ and $S_{2g}(\epsilon, \Theta, \tau)$ are the cross terms between the ground and excited states. Similarly, $S_{21}(\epsilon, \Theta, \tau)$ is the cross term between states $|1\rangle$ and $|2\rangle$, and represent the beat signal that is of our interest to determine the spectral phase of the XUV pulse. Although all cross terms, S_{1g} , S_{2g} , and S_{21} , are oscillating as a function of τ with beat frequencies corresponding to the energy differences of $4s-4p$, $4s-5p$, and $4p-5p$, respectively, we can extract the beat signal associated with the S_{21} term only through the frequency filtering. The reason why we do not choose the S_{jg} ($j = 1, 2$) terms to extract the spectral phase difference is that such a choice would complicate the analysis, because we must deal with the three partial waves for the photoelectron signals, p wave (photoelectrons from $|g\rangle$) in addition to the s and d waves (photoelectrons from $|1\rangle$ or $|2\rangle$). With the choice of the S_{21} term, instead, the partial waves we need to take into account are s and d waves only, and moreover, the d wave contribution to the S_{21} term can be eliminated by choosing the magic angle, $\Theta_0 = \cos^{-1}(1/\sqrt{3})$, for the photoelectron detection. At the magic angle detection, we can greatly simplify the expression of $S_{21}(\epsilon, \Theta, \tau)$ to be [15]

$$S_{21}(\epsilon, \Theta_0, \tau) = \frac{|C_1^* C_2|}{4\pi} \mathcal{R}_{s1} \mathcal{R}_{s2} \sqrt{I(\omega^{\text{xuv}} + \omega_{21}) I(\omega^{\text{xuv}})} \times \cos[\omega_{21}\tau + \phi_c + \Delta\phi_{21}(\omega^{\text{xuv}})], \quad (2)$$

in which C_k ($k = 1, 2$) are the complex probability amplitudes from state $|k\rangle$ after the two-color pumping, \mathcal{R}_{sk} is the radial component of the bound-free dipole moment from $|k\rangle$ to $|\epsilon s\rangle$, $\omega_{21} = \omega_2 - \omega_1$, and $\phi_c = \arg[C_1^* C_2]$. Now, $\Delta\phi_{21}(\omega^{\text{xuv}})$ is the spectral phase difference of the attosecond XUV pulse for the frequency interval of ω_{21} , i.e., $\Delta\phi_{21}(\omega^{\text{xuv}}) = \phi(\omega^{\text{xuv}} + \omega_{21}) - \phi(\omega^{\text{xuv}})$. In practice, $\Delta\phi_{21}(\omega^{\text{xuv}})$ can be experimentally determined by recording the photoelectron spectrum, $S_{21}(\epsilon, \Theta_0, \tau)$, as a function of τ and measuring the phase difference between the beat signal and the reference signal at every electron energy point ϵ . Note that the reference signal can be taken at any photoelectron energy, say, ϵ_0 , i.e., $S_{21}(\epsilon_0, \Theta_0, \tau)$, as long as the reference energy, ϵ_0 , is fixed throughout the entire measurement. This is so, since the spec-

tral phase, $\phi_{21}(\omega^{\text{xuv}})$, is a relative quantity. During the measurement, the atomic phase, ϕ_c , acts as a constant phase offset and would not affect the determination of the spectral phase difference, $\Delta\phi_{21}(\omega^{\text{xuv}})$. Once we have experimentally obtained $\Delta\phi_{21}(\omega^{\text{xuv}})$, we can retrieve the spectral phase of the XUV pulse, $\phi(\omega^{\text{xuv}})$, by the standard algorithm for the optical SPIDER [17]. In practice, the detection angle has a finite width around Θ_0 . In a recent paper we have shown that the spectral phase error introduced by the finite width of the detection around Θ_0 is rather small and can be safely neglected [15].

Now we investigate the influence of the noise on the accuracy of pulse reconstruction. Under the presence of noise, the total photoelectron signal can be expressed as

$$S = (1 + \sigma_I^{\text{xuv}}) \{A + B \cos[\omega_{21}\tau + \Delta\phi_{21}(\omega^{\text{xuv}}) + \sigma_\phi]\} + \sigma_I^{\text{inst}}, \quad (3)$$

where A and B are constants, σ_ϕ is the phase noise in the photoelectron signal, σ_I^{xuv} is the fraction of the intensity noise in the photoelectron signal caused by the shot-to-shot variance of the XUV pulse intensity, and σ_I^{inst} is the instrumental noise of the detector. It is worth noting that in the expression of Eq. (3) the $4s$ related beating terms have been eliminated, while the beat from $4p-5p$ and the photoelectrons directly ionized from $4s$, $4p$, $5p$ by the XUV pulse stay.

In order to obtain a visible beat of S_{21} , a frequency filtering has to be performed on the noisy signal, Eq. (3). After this filtering process, the signal-to-noise ratio would be greatly improved, but still a small portion of the intensity noises stay. We use A' and $\sigma_I^{\text{inst}'}$ to express the remaining A and σ_I^{inst} , respectively. Note that σ_I^{xuv} represents a relative fluctuation, and hence, would not change after the filtering. In our previous work on the VUV SPIDER [15], we examined the influence of the phase noise σ_ϕ and found that it can be effectively suppressed by choosing the energy shear, ω_{21} , to be $\sim 10\%$ of the bandwidth of the XUV pulse. Since this argument is obviously valid for the present case as well, we now examine the influence of the intensity noise in the photoelectron signal. Since σ_I^{xuv} and $\sigma_I^{\text{inst}'}$ are random quantities, we can mathematically express them as $\sigma_I^{\text{xuv}} = (\sigma_0^{\text{xuv}}) \eta^{\text{xuv}}$ and $\sigma_I^{\text{inst}'} = \sigma_0^{\text{inst}'} \eta^{\text{inst}'}$, where σ_0^{xuv} and $\sigma_0^{\text{inst}'}$ are fluctuation amplitudes, and the parameters η^{xuv} and $\eta^{\text{inst}'}$ are random functions with mean 0 and variance 1. Then the photoelectron signal can be expressed as

$$S_{21} \simeq A' + B \left\{ \cos[\omega_{21}\tau + \Delta\phi_{21}(\omega^{\text{xuv}}) + \sigma_\phi] + \frac{(\sigma_0^{\text{xuv}} \eta^{\text{xuv}}) A' + \sigma_0^{\text{inst}'} \eta^{\text{inst}'}}{B} \right\}, \quad (4)$$

where the small change of the beat amplitude B has been omitted since it would not affect the phase extracting of the beat signal. We factorize the random functions, η^{xuv} and $\eta^{\text{inst}'}$, as

$$\eta^{\text{xuv}} = (-\sin \phi_B) \eta_0^{\text{xuv}}, \quad \eta^{\text{inst}'} = (-\sin \phi_B) \eta_0^{\text{inst}'}, \quad (5)$$

where $\phi_B = \omega_{21}\tau + \Delta\phi_{21}(\omega^{\text{xuv}}) + \sigma_\phi$. Since the parameter $(-\sin \phi_B)$ lies within the range of $[-1, 1]$, we can find that the new random functions, η_0^{xuv} and $\eta_0^{\text{inst}'}$, are approximately equivalent to η^{xuv} and $\eta^{\text{inst}'}$. Under the assumption that the

intensity noise terms, $\sigma_I^{\text{xuv}} A'$ and $\sigma_I^{\text{inst}'}$, are much smaller than B , the intensity noise can be brought into the phase term of the quantum beat as

$$\begin{aligned} S_{21} &= A' + B \left[\cos \phi_B - \sin \phi_B \frac{(\sigma_0^{\text{xuv}} A') \eta_0^{\text{xuv}} + \sigma_0^{\text{inst}} \eta_0^{\text{inst}'}}{B} \right] \\ &= A' + B \left(\cos \phi_B - \sin \phi_B \frac{\sigma_I^{\text{xuv}} A' + \sigma_I^{\text{inst}'}}{B} \right) \simeq A' \\ &\quad + B(\cos \phi_B \cos \sigma_\phi^I - \sin \phi_B \sin \sigma_\phi^I) \\ &= A' + B \cos(\phi_B + \sigma_\phi^I), \end{aligned} \quad (6)$$

where $\sigma_\phi^I = (\sigma_I^{\text{xuv}} A' + \sigma_I^{\text{inst}'})/B$ represents the phase noise originating from the intensity variance. Comparing Eq. (6) with Eqs. (1) and (2), we find that A' and B in Eq. (6) can be expressed as

$$A' \propto S_{gg} + S_{11} + S_{22}, \quad (7)$$

$$B = \frac{|C_1^* C_2|}{4\pi} \mathcal{R}_{s1} \mathcal{R}_{s2} \sqrt{I(\omega^{\text{xuv}} + \omega_{21}) I(\omega^{\text{xuv}})}. \quad (8)$$

Note that A' is not exactly the signal of the photoelectrons ionized directly by the XUV pulse but a small portion of them remaining after the frequency filtering. The magnitude of A' should be proportional to $(S_{gg} + S_{11} + S_{22})$. Clearly σ_ϕ^I becomes smaller if B and hence the product of C_1 , C_2 , \mathcal{R}_{s1} , and \mathcal{R}_{s2} is larger. Provided with the same amount of the remained instrumental noise, $\sigma_I^{\text{inst}'}$, for both two-color and VUV SPIDERS, $\sigma_I^{\text{inst}'}/B$ for the two-color SPIDER can be easily 4 orders of magnitude smaller than that of the VUV SPIDER since C_1 and C_2 for the two-color SPIDER are 2 orders of magnitude larger. As for the $\sigma_I^{\text{xuv}} A'/B$ term in σ_ϕ^I , provided that the intensity variance of the XUV pulse, σ_I^{xuv} , is identical for both SPIDERS, σ_ϕ^I becomes smaller if we can make the beat contrast, B/A' , larger. This, however, is a very difficult requirement for the VUV SPIDER, since most atoms stay in the ground state due to the inevitably weak VUV pump pulse, and hence S_{gg} , which contributes to A' , is significant. In contrast it is not a problem at all for the two-color SPIDER, since the ground state population can be greatly minimized by the two-color pump technique under realistic conditions.

3. NUMERICAL RESULTS AND DISCUSSION

To be more specific we now assume that the atoms initially in the ground state are pumped to a coherent superposition of $4p$ and $5p$ by the fundamental and its second harmonic of a Ti:sapphire laser with the 20 fs duration and the peak intensities of $1.7 \times 10^{10} \text{ W/cm}^2$ and $2.6 \times 10^{12} \text{ W/cm}^2$, respectively. In this paper, we further assume that the fundamental and its second harmonic simultaneously interact with the K atom, i.e., the time lag between them is zero, and calculate the population of each state by numerically solving the time dependent Schrödinger equation. Since the intensity of the second harmonic pulse is more than TW/cm^2 , we also include the ionization loss by the two-color pump pulse in our calculations. The results are shown in Fig. 2, from which we can see that the population of the ground state is greatly minimized while the populations of the excited states are sufficiently large for superposition. Note that the differences in the photo-

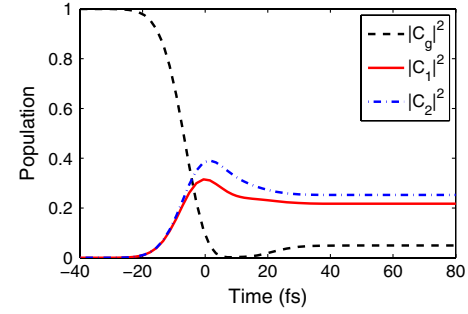


Fig. 2. (Color online) Population of the bound states as a function of time. The solid, dot-dashed, and dashed lines represent the populations of the $4p$, $5p$, and $4s$ states, respectively. The pump pulses have been assumed to have a duration of transform-limited 20 fs (peaked at 0 fs) with photon energies of 1.556 eV and 3.112 eV and peak intensities of $1.7 \times 10^{10} \text{ W/cm}^2$ and $2.6 \times 10^{12} \text{ W/cm}^2$, respectively.

emission timing of different quantum states [5], i.e., $4p$ and $5p$, are not included in our analysis, because the beat of the photoelectron signal is in the time scale of femtosecond, while the timing difference in the photoemission is only in the scale of attosecond. Based on the above qualitative arguments we can expect a much higher accuracy for pulse reconstruction with the two-color SPIDER.

Now we quantitatively compare the reconstruction accuracy of the VUV SPIDER and two-color SPIDER. We assume that the XUV pulse is slightly chirped from a transform-limited pulse with a duration of 300 as (6 eV spectral bandwidth for the full width at half-maximum) to a chirped pulse with a duration of 380 as, with a central photon energy at $\epsilon_0^{\text{xuv}} = \hbar\omega_0^{\text{xuv}} = 20 \text{ eV}$ and the spectral phase described by

$$\phi(\epsilon^{\text{xuv}}) = c_2(\epsilon^{\text{xuv}} - \epsilon_0^{\text{xuv}})^2 + c_3(\epsilon^{\text{xuv}} - \epsilon_0^{\text{xuv}})^3 + c_4(\epsilon^{\text{xuv}} - \epsilon_0^{\text{xuv}})^4, \quad (9)$$

where $c_2 = -3 \times 10^{-2} \text{ rad/eV}^2$, $c_3 = -1 \times 10^{-2} \text{ rad/eV}^3$, and $c_4 = 1 \times 10^{-3} \text{ rad/eV}^4$. To be more realistic we introduce a noise in the total photoelectron signals, as illustrated in Fig. 3. Briefly, we assume that the total instrumental noise and the total intensity noise of the XUV pulse, respectively, take

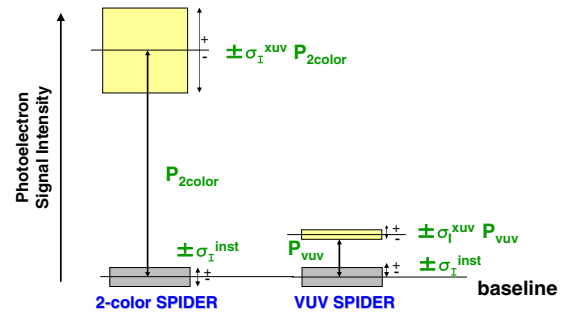


Fig. 3. (Color online) Schematic description of the noise introduced in the photoelectron signals that are used for the pulse reconstruction. We assume that the instrumental noise, σ_I^{inst} , and the intensity noise of the XUV pulse, σ_I^{xuv} , respectively, take the identical values for both two-color and VUV SPIDERS. The photoelectron signal, $P_{2\text{color}}$ or P_{vuv} , is proportional to the product of the bound state population and the XUV intensity, and $P_{2\text{color}}$ can be easily 4 orders of magnitude larger than P_{vuv} under the realistic experimental conditions as assumed in this work.

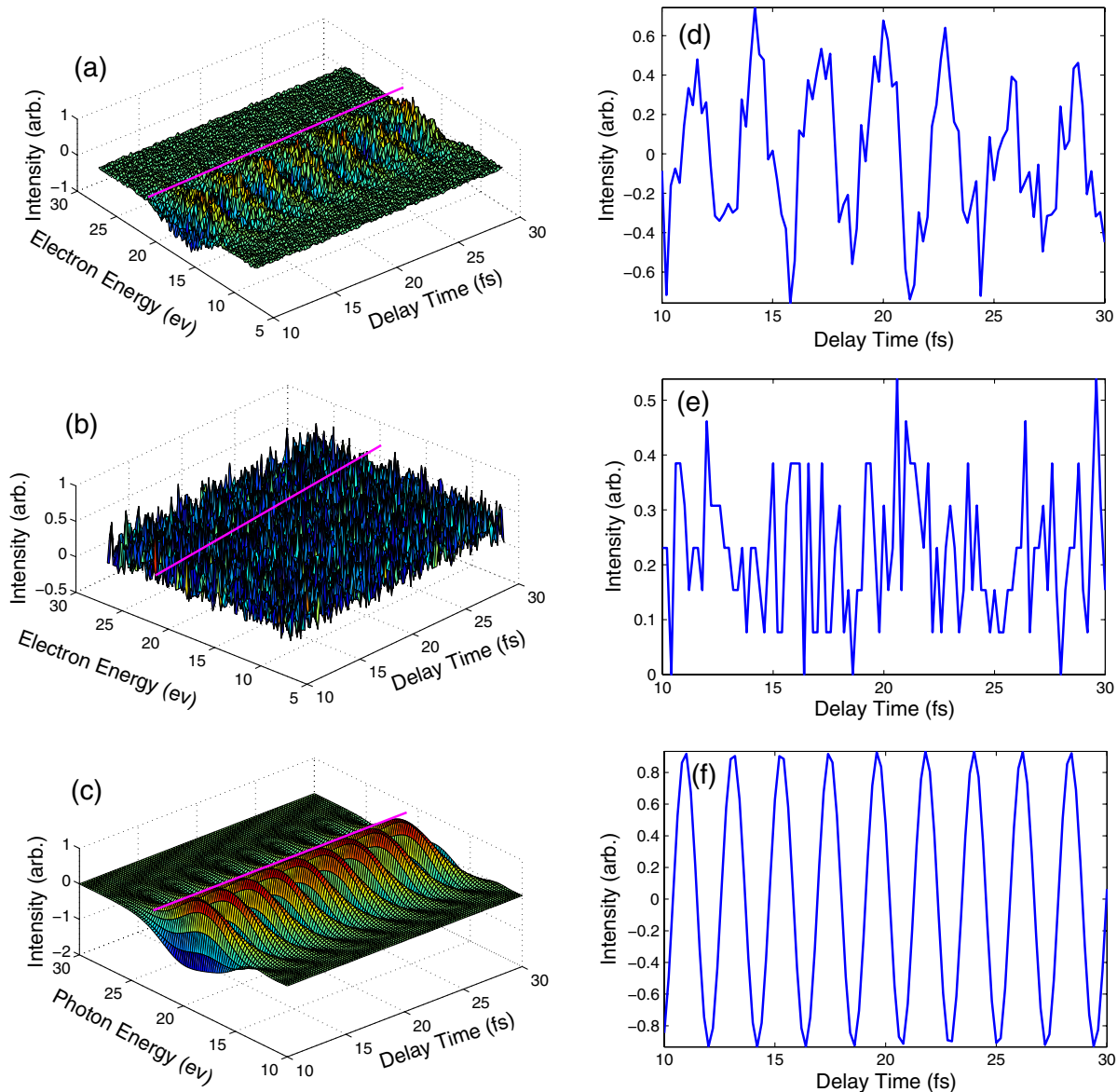


Fig. 4. (Color online) Noisy photoelectron spectra as functions of delay time and electron energy employed for the pulse reconstruction with the (a) two-color and (b) VUV SPIDERS. We also show the 1D cuts along the delay time at a photoelectron energy of 20 eV for the two SPIDERS, (d) two-color SPIDER, (e) VUV SPIDER. Graph (c) is the photoelectron spectra under the noise-free condition, and (f) is its 1D cut at 20 eV.

the identical values for both two-color and VUV SPIDERS, i.e., $\sigma_I^{\text{inst}} = 4.5 \times 10^{-5} I(\omega_0^{\text{xuv}})$ (in a.u.) and $\sigma_I^{\text{xuv}} = 20\%$, and the same frequency filtering method is used for both SPIDERS. For the two-color SPIDER the target is K atoms, and the parameters for the two-color pump pulse are identical to those used for Fig. 2. Note that the ionization process from $4s$, $4p$, and $5p$ by the two-color pump pulse has been included for the numerical evaluation of not only the population in those states (Fig. 2) but also the photoelectron signal. Namely, retrieval of the spectral phase of the specific beat signal we need, i.e., S_{21} , has been carried out by applying the frequency filtering for the beat signal, $S(\epsilon, \Theta, \tau)$, as already mentioned after Eq. (1). For the VUV SPIDER the target is H atoms, and the pump pulse has a photon energy of 11.3 eV with the 2 fs duration and the peak intensity of 10^{11} W/cm^2 . Under these conditions, the ionization probabilities for the two-color SPIDER is about 4 orders of magnitude larger than that of the

VUV SPIDER. The photoelectron spectra employed for the two-color and VUV SPIDERS are shown in Figs. 4(a), 4(b), and 4(e), respectively, which are to be compared with the noise-free photoelectron spectra shown in Figs. 4(c) and 4(f). From Fig. 4 we can see that the signal-to-noise ratio of the photoelectron spectra for the former is much better than that for the latter under the noisy environment. Note that the spectrum phase information is not directly extracted from these spectra data (Fig. 4). As we have described in Eq. (4), we need to perform Fourier transformation and choose the frequency component at ω_{21} as the target signal for reconstruction. Finally, Fig. 5 compares the reconstructed and original pulses by both SPIDERS, where ϵ_I and ϵ_ϕ , respectively, are the estimated temporal intensity and phase errors, according to Ref. [18]. From Fig. 5 it is clear that the two-color SPIDER offers a much higher reconstruction accuracy than the VUV SPIDER.

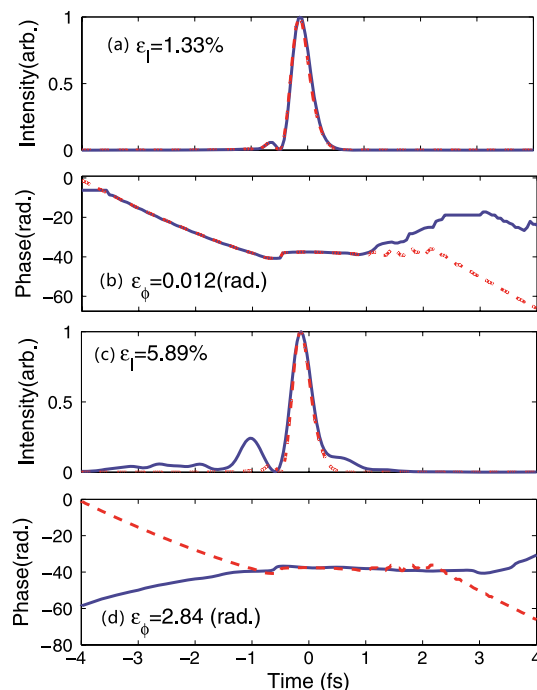


Fig. 5. (Color online) Comparison of reconstructed attosecond pulses by the (a) and (b) two-color SPIDER, and (c) and (d) VUV SPIDER. The solid and dashed lines represent the reconstructed and the original temporal profiles, respectively. ϵ_I and ϵ_ϕ are the estimated temporal intensity and phase errors, respectively.

4. CONCLUSION

In summary, we have proposed a photoelectron SPIDER using a two-color pumping. The K atom is used as a target, since its energy level structure best matches the two-color pumping by the fundamental (797nm) and its second harmonic of a Ti:sapphire laser. By measuring the phase offset of the quantum beat appearing in the energy-resolved photoelectron signal as a function of time delay between the two-color pump and XUV pulses, the spectral phase of the XUV pulse is accurately retrieved using the standard SPIDER algorithm. Compared with the VUV SPIDER [15], the two-color SPIDER has a remarkable advantage in terms of the reconstruction accuracy and the experimental feasibility due to the use of a Ti:sapphire laser. In contrast to the commonly used methods, such as FROG-CRAB and XSPIDER, which rely on SFA in one way or another, the two-color SPIDER, as well as the VUV SPIDER and Cs SPIDER, has a more transparent reconstruction process since they do not require any approximations.

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